Optical Pumping of Ruby by Laser Resonance Radiation-レーザー共鳴放射によるルビーの光ポンピング-

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論文内容要旨

A Introduction

Since introduction of the term "optical pumping" by Kastler[1], its meaning has broadened somewhat, eapecially with the advent of the quantum electronics era and the laser. Generally optical pumping is a method of creating and maintaining a non-thermal population distribution of atoms in different quantum states. While early experiments were concerned with optical pumping of gases with resonance radiation, more recently the technique has also been applied to solids. In particular broad band pumping is now commonly used to excite solid state lasers. In this thesis, the resonant optical pumping of the ruby transition $^4A_2 \rightarrow {}^2E$ (R_1 line) by a ruby laser beam is examined. The study is focussed on two applications of such a resonat interaction, (1) laser pumping of microwave masers and (2) self-Q-switching of ruby lasers. Specific objectives in (1) were to measure the laser induced change, δ, in the grouund state fine structure populations in ruby and to compare these with available theories and to construct new theoretical models where necessary. In particular hole-burning was incorporated into the theory for the first time and a new experimental technique introduced which directly demonstrates the inhomogeneous nature of the ruby R_1 line at low temperatures. In (2) a new teahnique of laser Q switching is described and good agreement between saturable absorber theory and experiment demonstrated.

B Laser-Pumped Ruby Maser

There are two primary advantages of optical pumping of microwave masers as first outlined by Hsu and Tittel[2]. in the case of CW pumping, (1) low noise maser operation is possible at bath temperatures much higher than the usual 42^0K value which is required when microwave pumping is used and (2) maser operation can be extended to higher frequenies. In the case of ruby at 78^0K say, the latter analysis shows that CW low noise maser operation is not possible, essentially because of a low ratio, f/w, of optical to microwave relaxation rate. An extension of the analysis into the time domain is introduced in Chapter B2.1 which shows that, under pulsed conditions, low noise operation becomes possible during a time approximately equal to the microwave relaxation time following application of the pump. The theory was verified experimentally using the apparatus shown in Fig.1. A spin temperature $T_s = -12\,^{\circ}K$ was measured with ruby laser pumping at a microwave ruby temperature of $78\,^{\circ}K$. Since the maser noise temperature[3] $T_n \simeq |T_S|$, this demonstrated that low noise maser operation at high bath temperature was indeed possible[4,5] as predicted by the time dependent theory.

The feasibility of CW high frequency (300 GHz) operation of a ruby microwave maser using optical pumping is discussed in chapter B 2.2. Calculations indicate that the pump power requirements are quite reasonable ($\simeq 5$ Watts/cm^2). Another advantage of optical pumping is pointed out, namely that because of the spin selection rules, 3 level maser operation using only the ground state Zeeman levels is not possible at the high magnetic fields required for mm wave operation. However optical transitions to ^2E electronic state are allowed and in fact constitute the only way to achieve maser action in ruby at mm wavelengths.

In chapter B 3, a new technique (internal optical pumping) is described for overcoming an unfavourable f/w ratio by combining laser and maser action in the same crystal. Unlike the external pumping scheme of Hsu and Tittel,[2] internal pumping allows the attainment of a low $|T_s|$ in ruby, at $78\,^{\circ}K$ for example, under CW conditions. The calculated pump power for such operation in ruby however is probably impractically high and other materials such as F centres in alkali halides which have much longer microwave relaxation times at higher temperatures are suggested as being more promising.

C Optical-Microwave Double Resonance Studies of Ruby

Comparison of experiment and the simple theory of chapter B showed that an order of magnitude larger pump energy was required in practice than predicted by theory. Such a discrepancy had also been previously noted by Devor et al[6] and was ascribed to a lack of frequency match betweem the ruby laser and absorption in the microwave ruby. To test this idea,a careful measurement of the laser and microwave ruby line positions was undertahen using direct optical spectroscopy as well as an optical-microwave double resonance technique[7]. This technique and theoretical extensions arising from these experiments are summarized below.

In the theoretical extension,consideration is given to three main factors neglected in the simple theory. These are,the finite optical linewidth which results in pumping of all the ground state levels in varying degrees depending on the laser frequency,the effect of hole-burning in the inhomogeneously broadend R_1 line at low temperatures and finally the fact that the microwave ruby is optically thick, that is the pump power varies with distance in the crystal. The apparatus used is shown in shown in Fig.1 with a modification which allowed variation of the sample temperature from 4.2 to 100°K. The cavity resnance frequency was 9.4 GHz. When the microwava ruby is irradiated by the pulsed laser beam,a change in the cavity reflection coefficient occures due to a change,δ,in the population diffence in the energy levels. Experimental and theoretical values of this change are shown in Fig.2 for the $1/2 \leftrightarrow -1/2$ transition (magnetic field is parallel to the crystal axis) as a function of the crystal temperature. Since the optical energy level spacing is temperature depedent, δ goes through positive and negative values as the laser pumps the upper or lower microwave levels. Of particular interest are the zero crossing points at 46 ^{o}K and 87 ^{o}K at which points one of the two lasers lines lies midway between the optical transitions from the +1/2 and -1/2 $^{4}A_{2}$ levels. The measurement of these zero crossing points allowed a precise determination of the relative positions of the laser and absorption lines. These positions were in good agreement with direct measurements using a large(35 foot) grating spectrograph. The solid line in Fig. 2 is a theoretical calculation valid for low pump powers. Agreement with experiment is reasonable except at higher temperatures where spin-lattice ralaxation effects become important. At higher pump energies(x in Fig.2),the hole-burning theory agreed with experiment within 20% at 4.2 ^{o}K. However the large chage in δ which occurs in the temperature range 10 to 40 ^{o}K was quite unexpected,since in this range,negligible change in the absorption frequency occurs.[8] Factors such as temperature-dependent spectral diffusion and transparency effects were theoretically examined,however they were found to be too small to explain the results. This anomalous behaviour was finally explained as due to a thermalization effect in the ^{2}E levels which,under saturation conditions, allows additional ions to be pumped into the excited state. Calculations of this effect were in good quantitative agreement with experiment.[7]

D Laser Induced Fluorescence Line Narrowing in Ruby

An important conclusion of the previous chapter was that under optical saturation conditions,"hole-burning" occurs in the R_1 line when it is excited by a laser whose frequency width is less than the R_1 inhomogeneous linewidth. In other words,only those ions which are resonant with the laser will saturate,while the rest of the line is unaffected. The evidence however is indirect as indeed are other observations such as the temperature dependence of linewidth.[8] In this chapter,the inhomogeneous nature of the R_1 line at low temperatures is directly demonstrated using a technique of laserinduced fluorescent line-narrowing.[9] The apparatus used is shown in Fig.3. A liquid nitrogen cooled ruby laser resonantly excites the R_1 line,after which the fluorescence is frequency-analyzed by a high-resolution scanning Fabry-Perot interferometer. Initial results showed a narrowing from the inhomogeneous width of 2.2 GHz to a value of 600 MHz. More recent results[9] have resulted in values as low as 60 MHz,which is approaching the homogeneous width of $\simeq$ 10 MHz implied by photon-echo studies in zero magnetic field.[10] The latter width arises from the $Cr-Al^{27}$ electron-nuclear spin dipole interaction. An interesting aspect of the observations is that no spectral diffusion occurs for times up to 10 msec after excitation. These results support Birgeneau's[11] recent calculations which show that earlier postulated multipolar interactions do not produce single-ion to single-ion energy transfer in concentrated ruby but rather that the interaction must be of a short range type such as exchange.

E Self-Q-Switching of Ruby Lasers

A novel technique for producing high power optical pulses from a ruby laser is described.[12] It is shown that when Cr ions in an unpumped portion of a ruby laser rod resonantly interact with laser radiation produced by the pumped portion of the rod, then rapid saturation of the unpumped ions occurs resulting in a corresponding rapid decrease of the optical cavity loss. As is well known, such a loss switching mechanism in lasers results in a form of operation called Q-switching. In the present case, the term self-Q-switching is introduced since the usual optical switch such as a Kerr or Pockel cell external to the laser rod is not required. An essential feature of the self-Q-switching mechanism is that a non-uniform cavity photon density is required to obtain saturable absorber switching[13] by unpumped laser atoms. This was achived by the use of a rooftop laser rod in which the photon density is doubled in the volume contained in the rooftop because of folding of the beam on itself. When the rooftop was shielded from the pump, a Q-switched laser pulse of peak power 200 KW and width 6 nsec was produced. This operation only occurred at liquid nitrogen temperatures, ceasing at temperatures above 150°K. The Q-switching behaviour and its temperature dependence are quantitatively explained by saturable giant pulse theory.[13] A plot of theoretical and experimental pulse shapes is shown in Fig.4. The theoretical curve was adjusted to the experimental points by variation of the theoretical parameters. A comparison of latter parameters with values deduced from experimental conditions showed good agreement.[12]

F Conclusions

The main conclusions and accomplishments of this work are listed below.

(1) Theoretical prediction that laser pumping of microwave masers could produce spin temperatures(absolute value) much less than the bath temperature was experimentally verified. For a ruby system,a spin temperature of $-12^{o}K$ was measured[4,5] at a bath temperature of $78^{o}K$.

(2) A difficulty which arises with ruby is that at higher temperatures,the optical to microwave relaxation time ratio becomes unfavourable for CW maser action using optical pumping. A method of circumventing this problem was suggested and a detailed theory outlined.

(3) Quantitative comparison between theory and experiment of the effects of laser pumping on the ground state populations of ruby showed that several factors not considered in previous work[6] required consideration. The most important of these were line-overlap,hole-burning and thermalization in the ^{2}E levels. In addition a double-resonance technique was developed to precisely locate the laser frequeocy relative to the absorption frequencies.[7]

(4) A limiting factor in the resolution of sharp lines in gases and solids by conventional spectroscopy is inhomogeneous broadening. Doppler inhomogeneous broadening in gases has recently been overcome by a laser saturation technique.[14] In this thesis a laser technique was described which,for the first time has overcome inhomogeneous broadening effects in a solid.[9]

(5) A new technique of Q-switching ruby lasers was reported which involved saturation of an unpumped portion of the ruby rod.[12]

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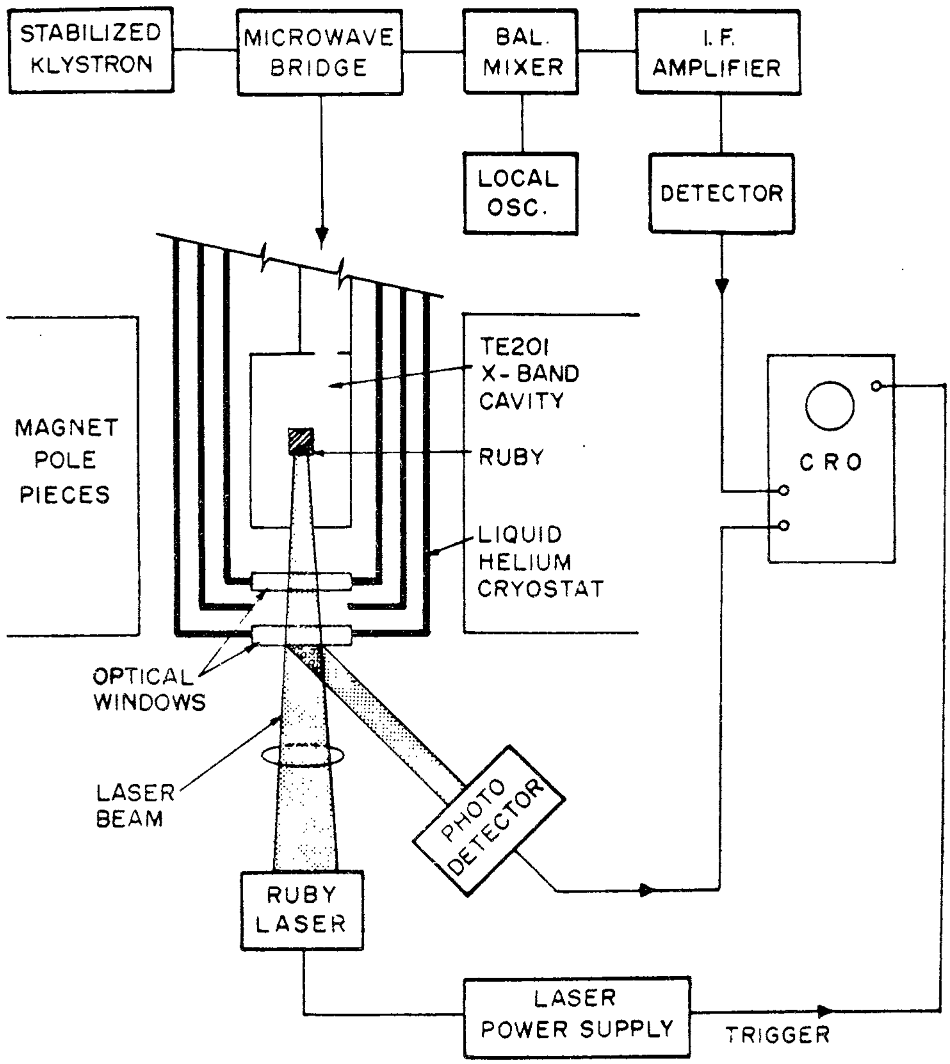

Fig.1 Schematic of apparatus used for experiments on laser-pumped maser and optical microwave double resonance studies of ruby.

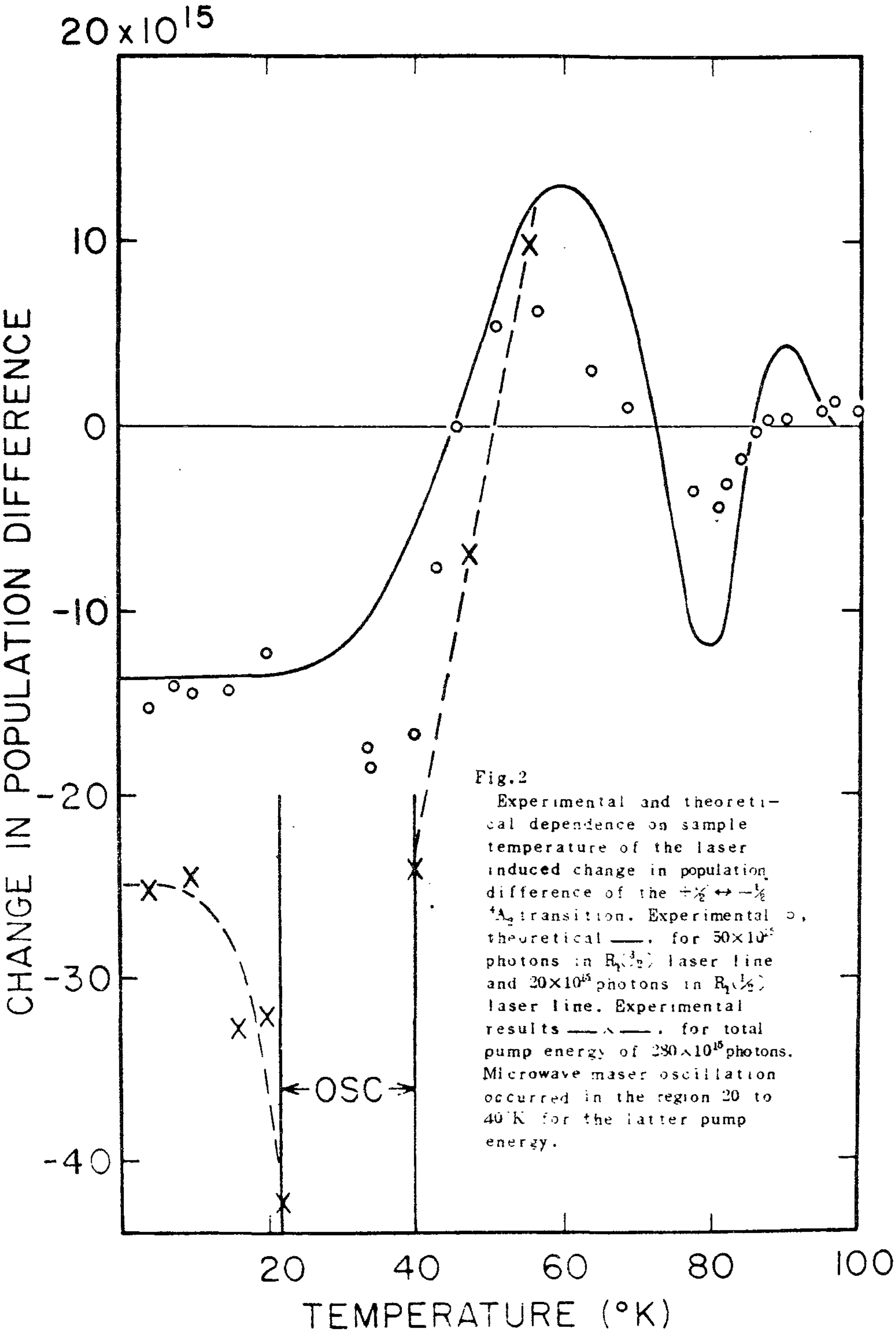

Fig.2
Experimental and theoretical dependence on sample temperature of the laser induced change in population difference of the $+\frac{1}{2} \leftrightarrow -\frac{1}{2}$ $^4A_2$ transition. Experimental ○, theoretical ——, for 50×10^{15} photons in $R_1(\frac{3}{2})$ laser line and 20×10^{15} photons in $R_1(\frac{1}{2})$ laser line. Experimental results —×—, for total pump energy of 280×10^{15} photons. Microwave maser oscillation occurred in the region 20 to 40°K for the latter pump energy.

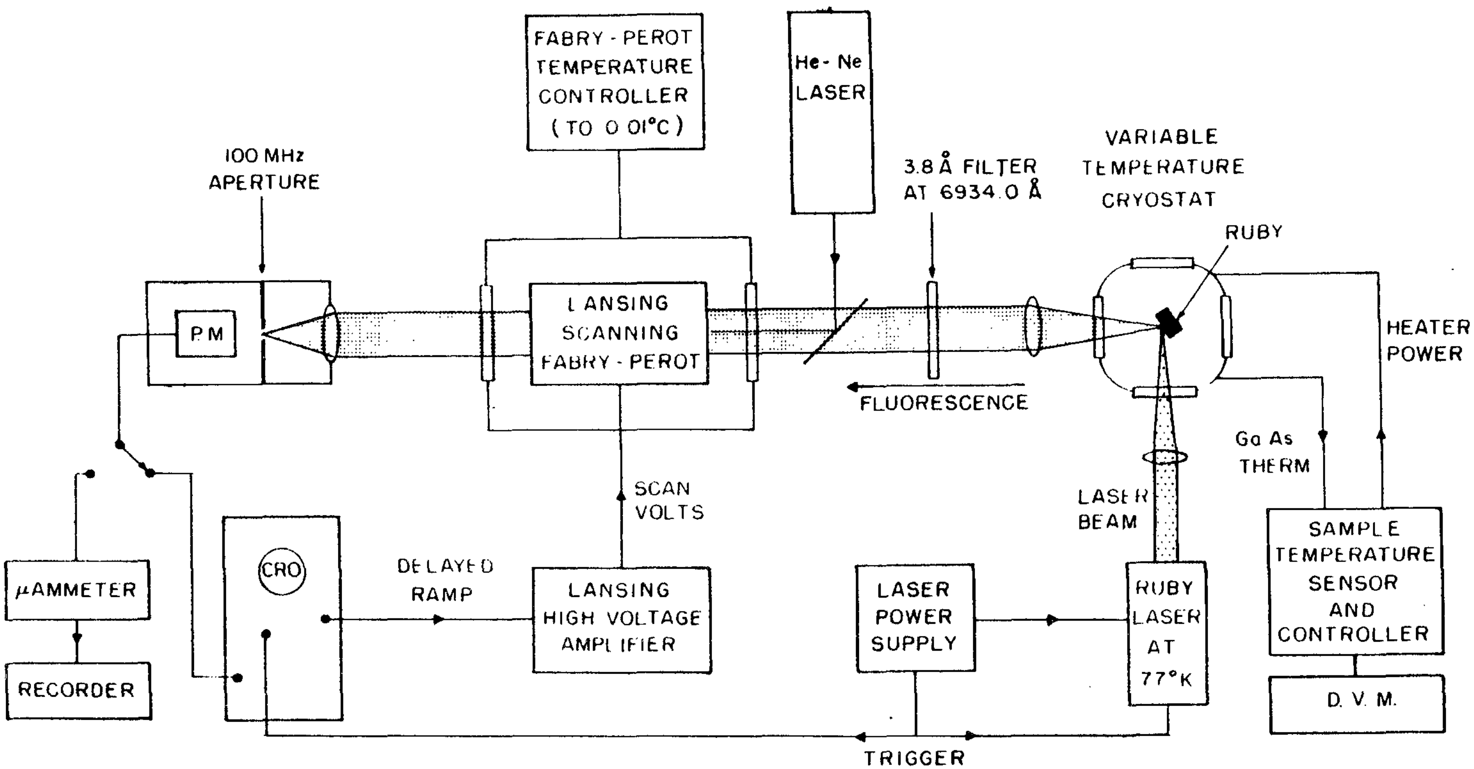

Fig.3 Schematic of apparatus used for laser induced fluorescent line narrowing experiments.

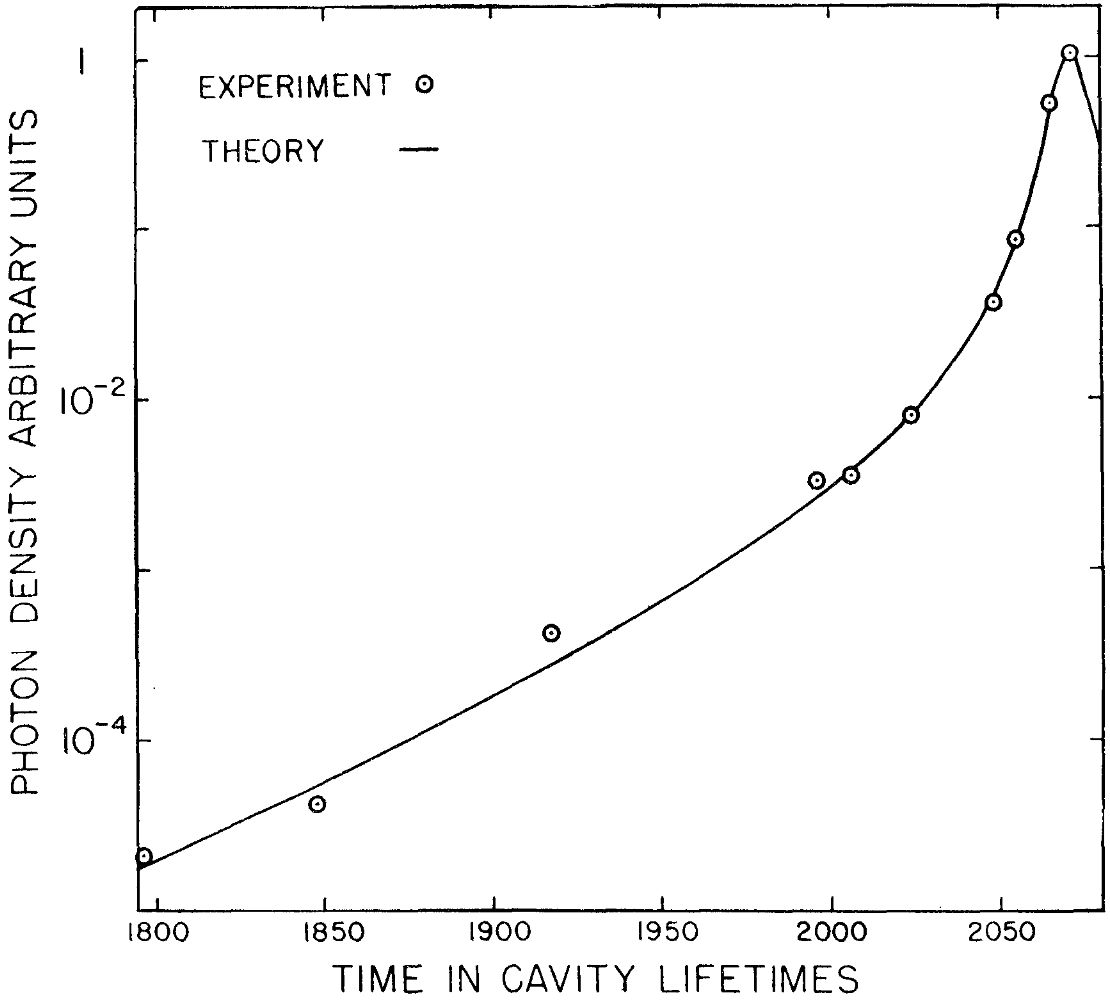

Fig.4 Experimental and theoretical time development of self -Q- switched laser pulse.

審査結果の要旨

近年におけるレーザー工学の急速な進展に伴い，さまざまの新しい現象や応用が見出され，広汎な実用化が進められている。この間レーザー光の時間的ならびに空間的な基本特性や高輝度特性などが明らかになったが，従来の光にくらべてはるかにすぐれた単色性を有するレーザー光に共鳴する物質が，そのレーザー光によって励起された場合，どのような応答を示すかについては従来ほとんど系統的研究が行なわれていなかった。本研究はその端緒をひらくのに貢献した基礎的研究と幾つかの工学的応用に関するもので，レーザー材料としてよく知られたルビー結晶の，ルビー・レーザー光による光ポンピングによって生ずるいろいろな効果をマイクロ波と光波の両領域にわたって綿密に解明したものである。本論文は全文6章よりなる。

第1章は序論で，本研究の目的の位置づけと要点を述べたものである。

第2章はルビー・レーザー光で光ポンピングされたルビーのマイクロ波メーザーを，増幅器としての低雑音特性の観点から理論的ならびに実験的に検討したものである。特に，光ポンピングによって結晶自体の冷脚温度よりも十分低い雑音温度が実現可能であることをはじめて実証したものであって，理論との良好な一致を示している。

第3章はルビー中のクロム・イオンのレーザー光とマイクロ波に対する二重共鳴現象についての詳細な研究結果を述べたものである。ここではルビーの吸収線と，光ポンピスグを行なうレーザー光との両者の波長関係が本質的な役割を果すことを適確に把握し，その定量的解析にもとづいて，スペクトル線の重なり効果や不均一な拡がりの影響，さらに基底準位内のクロス緩和現象などの二重共鳴への寄与の仕方を統一的に解明している。これらの成果は本論文の基礎をなすものであり，すぐれた知見である。

第4章は第3章の成果を足場として，さらにルビー・レーザー光によって照射されたルビー結晶から生ずる螢光スペクトルは，その線幅が狭くなっていることをはじめて実験的に見出した結果を述べ，それに考察を加えたものである。この事実は結晶の欠陥や転位などの不完全性に起因するスペクトル線の不均一な拡がりの中から，均一な拡がり幅の情報を定量的に導くための新しい手段を提供するもので，光物性的にも，またレーザー工学の立場からも極めて重要なものである。

第5章は低温に冷却されたルビー・レーザーで独立に設けられた変調素子を用いずに，Qスイッチを行なう新しい方法を実験によって実証するとともに，理論的解釈の妥当性をたしかめたものである。

第6章は総括と結言である。

以上要するに，本論文は高輝度で単色性にすぐれたルビー・レーザー光を，レーザー材料とは

別に設けられたルビー結晶に照射して光ポンピングを行なうことにより生ずる共鳴効果を詳細に論じ，その効果を通してマイクロ波ならびに光波領域において得られるメーザー増幅作用や螢光線幅のせばめ効果，Qスイッチ発振などの解明のための基礎を与えたばかりでなく，多くの新しい知見を加えたものであって，量子電子工学へ寄与するところが少なくない。

よって，本論文は工学博士の学位論文として合格と認める。